

HYPERFINE INTERACTIONS IN ANTIFERROMAGNETIC EuTe USING THE Te-125 MOSSBAUER RESONANCE

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ABSTRACT

Europium telluride crystallizes in the NaCl structure and is antiferromagnetic with a Néel temperature T_N of 9.6 K. We have used the 35.5 keV Mössbauer transition in Te-125 to examine the nature of the spontaneous magnetic moment which has been observed in conducting samples below T_N . At 80 K the Mössbauer resonance spectrum consisted of a single absorption line comparable in width with the spectrum of nonmagnetic cubic ZnTe, indicating the absence of a significant quadrupole splitting at this temperature. At 4.2 K the line-width increased by a factor of 1.5, which is equivalent to a single magnetic hyperfine field at the Te sites of about 73 kOe. The line showed gradual broadening with increasing external longitudinal magnetic field up to 70 kOe at 4.2 K. These observations are consistent with the magnetization measurements as well as with recent spin-echo NMR experiments showing resonances which may be attributed to Te-125. We conclude from the shape of the Mössbauer line that the broadening observed at 4.2 K is most likely due to a single magnetic hyperfine field of magnitude 73 kOe acting at the Te sites. An applied magnetic field $H < 65$ kOe induces an internal field component of opposite sign and nearly equal magnitude at the tellurium site, but near the canted to paramagnetic transition ($H \approx 66$ kOe at 4.2 K) the observed hyperfine field increases more rapidly with H , reaching a value of 112 kOe when $H = 70$ kOe.

INTRODUCTION

The divalent europium monochalcogenides (EuO, EuS, EuSe, and EuTe) crystallize in the f.c.c. NaCl structure and are magnetically ordered at low temperature as a result of the localized spins on the Eu^{2+} ions. At sufficiently low temperature EuO, EuS, and EuSe are ferromagnetic, while EuTe is antiferromagnetic. The relatively simple lattice structure and variety of magnetic phenomena observed in this class of materials have resulted in their properties being extensively investigated in recent years.^{1,2}

Samples of EuTe prepared by two different methods were previously examined by magnetization³ and spin-echo NMR⁴ techniques. Insulating samples ($\rho \approx 10^6$ ohm-cm) behaved like pure stoichiometric EuTe, exhibiting ideal antiferromagnetic behavior at an external magnetic field $H = 0$ for $T < T_N$.³ Conducting samples ($\rho \approx 10^{-2}$ ohm-cm) were found to have a spontaneous magnetic moment at $H = 0$ for $T < T_N$.³ Both types exhibited NMR spectrum echoes which were tentatively identified as Te-125 and Te-123 resonances.⁴ The magnetization and NMR results are consistent in that they both suggest, for the conducting sample, and possibly for the non-conducting sample also, a departure from the magnetic symmetry imposed by the NaCl structure.

Oliveira, et al.³ and Shapira, et al.⁵ have speculated that the anomalous magnetic behavior of the conducting samples is due to the magnetic effects of conduction electrons which are present because of lack of perfect stoichiometry in the sample, voids, impurities, etc. These effects should be nonlocalized and uniformly characteristic of the entire sample. It is the purpose of the present experiment to investigate this hypothesis

and to make contact with the NMR results of Raj, et al.⁴

EXPERIMENTAL RESULTS

The EuTe Mössbauer absorber was prepared from pieces of the same crystal (#102) used in the magnetization³ and NMR⁴ experiments by grinding the EuTe together with fine powdered alumina under dry nitrogen, and hermetically encapsulating the mixture in a plastic disc. The absorber thickness was approximately 3 mg/cm² Te-125. The reference single-line absorber, containing 1.5 mg/cm² Te-125, was similarly prepared, using chips from a single crystal of cubic ZnTe.⁶

Mössbauer spectra⁷ of EuTe (#102) and cubic ZnTe were obtained at 80 K for an external magnetic field $H = 0$, and at 4.2 K for several values of H from 0 to 70 kOe. Examples of the data are shown in Fig. 1, where least squares fit single Lorentzian solid lines are shown superimposed on the data points. All the spectra have the appearance of single lines of various widths, showing no resolvable structure, and having a nearly Lorentzian shape. Single line least square fits were used for the purpose of comparing ZnTe spectra in a known magnetic field with the EuTe spectra. Typical least square fits are shown in Fig. 2 and the results are summarized in Table I. The ZnTe and EuTe absorbers are not quite the same effective thickness and the calculation of the internal magnetic field, H_{int} , at the Te site in EuTe takes this into account. The line shape of the broadened EuTe sample (4.2 K, $H = 0$) matches that observed in ZnTe (4.2 K, $H = 70$) with an accuracy which strongly favors an interpretation of the broadening in EuTe as due to a single internal field, $H_{\text{int}} = 73$ kOe.

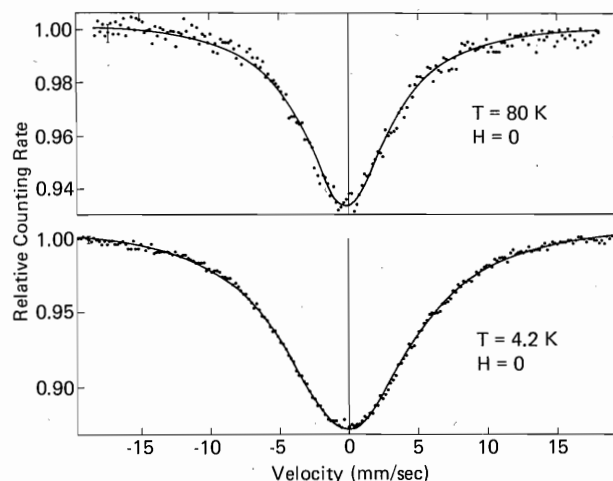


Fig. 1. Spectra of EuTe (#102) showing data points together with least squares single line Lorentzian fit. The counting statistical error is shown by the bar near the left background in each spectrum.

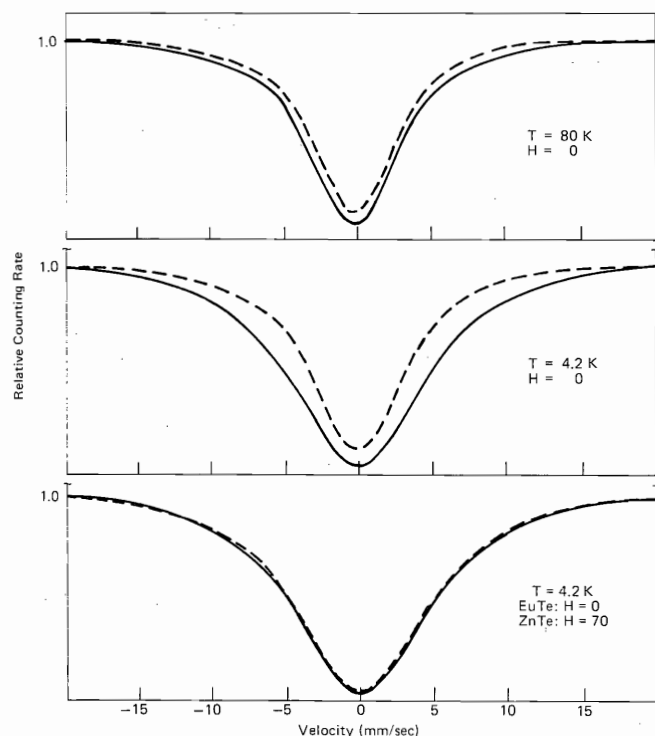


Fig. 2. Spectra showing superimposed least squares fits of EuTe (#102) (solid line) and ZnTe (dashed line) for various temperatures and magnetic fields. The data points were omitted to avoid cluttering the figure.

Table I. Summary of experimental results. I and Γ are, respectively, intensity and full width at half maximum intensity of least square fit Lorentzian lines.

		ZnTe			EuTe # 102		
T	H	I	Γ	H_n	I	Γ	H_n
Kelvin	kOe	%	mm/sec	kOe	%	mm/sec	kOe
80	0	3.9	6.80	0	6.8	7.50	0
4.2	0	13.0	7.70	0	17.0	11.20	73
4.2	50	12.5	9.60	50	18.9	11.40	78
4.2	65	12.2	10.10	65	18.9	11.50	81
4.2	70	12.0	10.30	70	18.9	12.70	113

DISCUSSION

The NMR results of Raj, et al.⁴ identify two echoes near 115.1 and 98.0 MHz, with the sample at 1.4 K, as having the proper frequencies and intensity ratio to be consistent with Te-125 and Te-123 resonances in a field $H_{int} = 86$ kOe. The width and shape of the EuTe Mössbauer line in zero external field at 4.2 K (see Table I) is consistent with a single hyperfine field $H_{int} = 73$ kOe at the Te sites. For $T_N = 9.6$ K a sublattice magnetization curve, approximated by a Brillouin function with $S = 7/2$, indicates that $M(4.2K) \approx 0.85 M(1.4K)$.⁸ The NMR result therefore corresponds to $H_{int} \approx 73$ kOe by the Mössbauer measurements, if we make the reasonable assumption that the net internal magnetic field (everywhere, and at the Te site in particular) follows the sublattice magnetization. Because the Mössbauer line components are unresolved, it is not possible to eliminate the possibility of a small quadrupole interaction or a non-unique value of H_{int} . However, the observed line shape, compared with ZnTe in an external field, discriminates against a field dis-

tribution that is weighted towards low values. The excellent agreement between the Mössbauer and the NMR results supports the hypothesis that the field is unique, or at least that it has a major component that is unique. The NMR results taken together with the Mössbauer results indicate that all, or nearly all, the Te nuclei in the sample experience the hyperfine field, and not just nuclei in or near domain walls.

It is possible that there may be a small distortion of the crystal structure coincident with the magnetic phase transition at $T = T_N$. Such behavior has been observed in other rare-earth monochalcogenides⁹; although in this case Eu^{++} is a S-state ion which is believed to couple only weakly to the lattice. Our inability to obtain a satisfactory fit to the $T = 4.2$ K, $H = 0$ spectrum with a quadrupole doublet, together with the NMR results, supports the view that a possible crystal distortion does not produce a significant quadrupole broadening compared with the magnetic hyperfine broadening.

In an applied magnetic field, the observed field, H_n , increases in magnitude, but by much less than H , until $H \approx 65$ kOe, where the AF \rightarrow P magnetic phase transition occurs.³ $H_n = H + H_{DM} + H_{int}$ where H_{DM} is the demagnetization field. For the sample geometry used (a thick disc filled with separated, irregularly-shaped EuTe polycrystals), $H_{DM} \approx 3.0$ kOe. The results (see Table I) support a model in which there is an initial contribution to H_{int} , negative with respect to H , which changes more rapidly in the vicinity of the AF \rightarrow P transition.

There is still no direct evidence for a lattice distortion which would reduce the Te site symmetry. The origin of H_{int} at the Te sites remains unknown; the NMR results indicate that it is present in both conducting and non-conducting samples.

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